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(54) Process and reactor for carrying out non-adiabatic catalytic reactions

(57) Process for carrying out non-adiabatic reactions comprising the steps of:

introducing in parallel a first stream of reactants into a first reaction space and a second stream of reactants into a second reaction space;
at reaction conditions contacting the first reactant stream with a catalyst in the first reaction space in indirect heat exchange with a heat exchanging medium and contacting the second reactant stream with a catalyst in the second reaction space in indirect heat exchange with a heat exchanging medium, and withdrawing a first and second stream re-

formed product gas; and
the catalyst in the first reaction space being arranged within a tubular reactor in indirect heat exchanging relationship with the heat exchanging medium by introducing the medium into tubular heat exchange space concentrically surrounding the tubular reactor with the first reaction space, the catalyst in the second reaction space being arranged on shell side of a heat exchange space in indirect heat exchanging relationship with the heat exchanging medium.

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Description

[0001] The present invention relates to a process and reactor system for carrying out non-adiabatic reactions proceeding in a process gas in presence of a catalyst exothermically or endothermically in indirect heat exchange with an appropriate heat exchange medium.

[0002] A general object of this invention is thus to provide a process for carrying out non-adiabatic reactions comprising the steps of:

introducing in parallel a first stream of reactants into a first reaction space and a second stream of reactants into a second reaction space,

at reaction conditions contacting the first reactant stream with a catalyst in the first reaction space in indirect heat exchange with a heat exchanging medium and contacting the second reactant stream with a catalyst in the second reaction space in indirect heat exchange with the heat exchanging medium, the catalyst in the first reaction space being arranged within a tubular reactor in indirect heat exchanging relationship with the heat exchanging medium by introducing the medium into tubular heat exchange space concentrically surrounding the tubular reactor with the first reaction space, the catalyst in the second reaction space being arranged on shell side of a heat exchange space in indirect heat exchanging relationship with the heat exchanging medium.

[0003] The invention is in particular useful in carrying out steam reforming reactions in a hydrocarbon feed stock by heat supplied from hot effluent gas from an autothermal steam reforming reactor and steam reformed product gas from the process.

[0004] A specific embodiment of the reaction system according to the invention is described more detailed in the following description by reference to the drawings in which Fig. 1 shows schematically a reaction system being used in the production of a gas with a high content of hydrogen and/or carbon monoxide from steam-reforming of a hydrocarbon feed stock.

[0005] Steam reforming is an endothermic chemical reaction, where hydrocarbons and steam react on a steam reforming catalyst, and if appropriate heat is supplied to the location of the reaction.

[0006] The reactor system being used in this embodiment consists of three reactors, wherein the steam reforming process is carried through. The three reactors R1, R2 and R3 are operated in parallel.

[0007] R1 is an adiabatic reactor. The reactants for the process in R1 consist of hydrocarbon, steam and an oxygen rich gas being introduced into the reactor at an appropriate temperature and mixed. The oxygen and the hydrocarbon will react by combustion and result in a hot gas of residual hydrocarbon, steam and resulting

in products from the combustion. Subsequently, the hot gas is passed through a bed of reforming catalyst and catalytically converted to a hot mixture of hydrogen, carbon monoxide and carbon dioxide.

[0008] R2 and R3 are two plug flow reactors. The reactants for the process in R2 and R3 are a mixture of hydrocarbon and steam, which is heated to an appropriate temperature before flowing through a bed of reforming catalyst. Walls surround and enclose the catalyst beds of R2 and R3. A hot gas is flowing outside these walls countercurrent to the reacting gases in the catalyst beds. Heat is conducted through the walls from the hot gas to the reacting gases, while the gases are converted to a hot mixture of hydrogen, carbon monoxide and carbon dioxide.

[0009] The product gases from R1, R2 and R3 are mixed and form the hot gas flowing outside the walls of R2 and R3, where they form the heat source of the reactions in R2 and R3. This gas is called the heating gas.

[0010] As a general advantage of the invention, the walls of R2 and R3 can be arranged in a layout so as to form an optimal channel for the heating gas.

[0011] The invention provides, furthermore, a reaction system being in particular useful for carrying out the above process. In general, the reaction system of this invention comprises connected in parallel a first and a second reaction compartment being adapted to hold a catalyst and to receive a reactant stream, the first compartment being in form of a reactor tube, wherein

[0012] a first heat exchange space concentric and spaced apart surrounds the first reaction compartment, and the second reaction compartment surrounds a second heat exchange space.

[0013] Reactor R2 contains the catalyst inside tubes. Reactor R3 holds the catalyst outside the tubes. The combined reactor R2 and R3 comprises a number of double-tubes, where the inner tubes are catalyst filled (R2) and the double-tubes are in addition arranged in a pattern allowing the volume between the double-tubes to be filled with catalyst as well, i.e. reactor R3. The sensible heat from the combined product gas from the reactors R1, R2 and R3 is cycled back to the reactors R2 and R3. The product gas is flowing in annular channels provided by the double-tubes, countercurrently to the flow in the reactors R2 and R3. Heat is supplied to reactor R2 via the inner wall of the double pipes and the reactor R3 is supplied with heat from the outer wall of the double-tubes.

[0014] The advantage of the combined reactor as shown in Fig. 2 is that the heat exchange channels are utilised in an optimal manner, i.e. both the inner wall and the outer wall are utilised as exchange heat surfaces thus making optimal use of expensive material. This also leads to a very compact design of equipment compared to other types of heat exchange reformers and at the same time provides low pressure drop.

[0015] On cooling the product gas, a certain risk of metal dusting corrosion exists. A further advantage of

the combined reactor design is restricted risk of metal dusting to a limited surface.

[0016] The double tube dimensions are typically: Inner tube OD 50 to 140 mm and outer tube OD 80 to 170 mm. The layout can be but need not be arranged in such a way that the heat exchange/ area/catalyst volume ratio is equal for the inner tubes and the outer tubes.

second reaction compartment are arranged within a common shell.

- 5 6. Reaction system of claim 4, wherein the first and second heat exchange space are formed by a common passageway.

Claims

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1. Process for carrying out non-adiabatic reactions comprising the steps of:

15 introducing in parallel a first stream of reactants into a first reactions space and a second stream of reactants into a second reaction space;
at reaction conditions contacting the first reactant stream with a catalyst in the first reaction space in indirect heat exchange with a heat ex-
20 changing medium and contacting the second reactant stream with a catalyst in the second reaction space in indirect heat exchange with a heat exchanging medium, and withdrawing a first and second steam reformed product gas;
25 and
the catalyst in the first reaction space being arranged within a tubular reactor in indirect heat exchanging relationship with the heat exchanging medium by introducing the medium into
30 tubular heat exchange space concentrically surrounding the tubular reactor with the first reaction space, the catalyst in the second reaction space being arranged on shell side of a heat exchange space in indirect heat exchanging re-
35 lationship with the heat exchanging medium.

2. Process of claim 1, wherein the non-adiabatic reaction is endothermic steam reforming of a hydrocarbon feedstock. 40
3. Process of claim 1, wherein the heat-exchanging medium comprises an effluent stream from autothermal steam reforming of a hydrocarbon feedstock and/or the product gas. 45
4. Reaction system for carrying out non-adiabatic catalytic reactions, comprising connected in parallel a first and second reaction compartment being adapted to hold a catalyst and to receive a reactant stream, the first compartment being in form of a reactor tube, wherein
50 a first heat exchange space concentric and spaced apart surrounds the first reaction compartment, and the second reaction compartment surrounds a second heat exchange space. 55
5. Reaction system of claim 4, wherein the first and

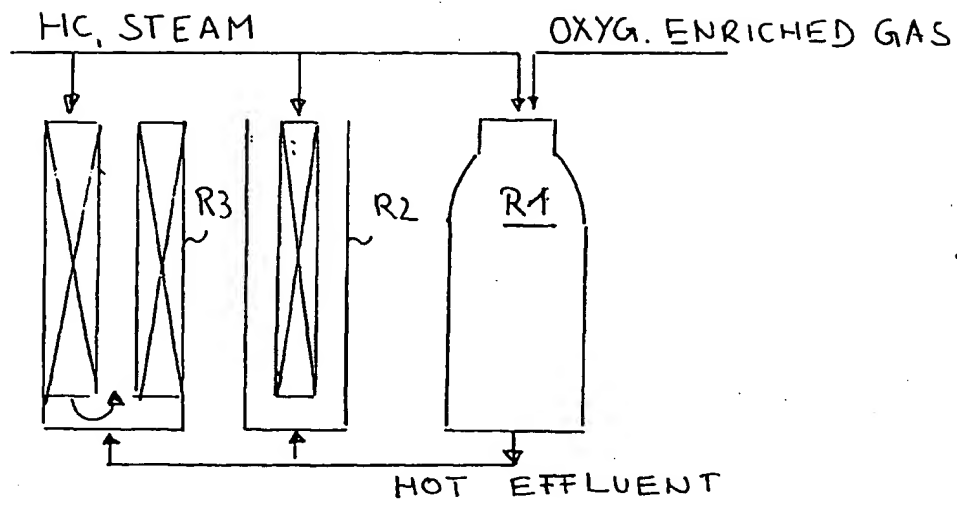


Fig. 1

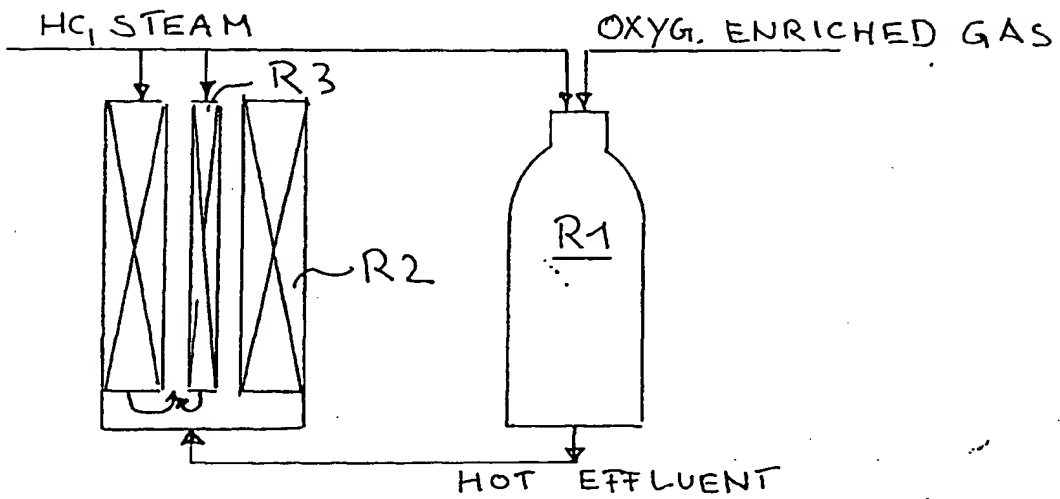


Fig. 2



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EUROPEAN SEARCH REPORT

Application Number
EP 00 12 5077

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
X	EP 0 271 299 A (BRITISH PETROLEUM CO PLC) 15 June 1988 (1988-06-15) * the whole document *	1,2,4,5	C01B3/38 B01J8/02 B01J8/06
X	PATENT ABSTRACTS OF JAPAN vol. 008, no. 187 (C-240), 28 August 1984 (1984-08-28) & JP 59 083904 A (TOUYOU ENGINEERING KK), 15 May 1984 (1984-05-15) * abstract *	1,4	
A	EP 0 334 540 A (ICI PLC) 27 September 1989 (1989-09-27) * the whole document *	1,4	
A	US 4 079 017 A (BECKER COLMAN LEE ET AL) 14 March 1978 (1978-03-14) * the whole document *	1,4	
A	EP 0 326 662 A (UHDE GMBH) 9 August 1989 (1989-08-09) * column 3, line 43 - column 5, line 33 * * figure 3 *	1,4	
A	EP 0 440 258 A (TOPSOE HALDOR AS) 7 August 1991 (1991-08-07) * page 4, line 25 - page 5, line 15 * * figures 1,2 *	1,4	
A	US 3 958 951 A (KUO CHI SHENG ET AL) 25 May 1976 (1976-05-25) * column 5, line 16 - column 6, line 7 * * figure 2 *	1,4	
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 7 February 2003	Examiner Van der Poel, W
<p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document</p>			

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**ANNEX TO THE EUROPEAN SEARCH REPORT
ON EUROPEAN PATENT APPLICATION NO.**

EP 00 12 5077

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on
The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

07-02-2003

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
EP 0271299	A	15-06-1988	AU 598074 B2	14-06-1990
			AU 8223287 A	16-06-1988
			CA 1329704 A1	24-05-1994
			DE 3771044 D1	01-08-1991
			EP 0271299 A2	15-06-1988
			JP 63218792 A	12-09-1988
			NO 875141 A , B,	13-06-1988
			NZ 222835 A	28-08-1990
			ZA 8709257 A	30-08-1989
JP 59083904	A	15-05-1984	NONE	
EP 0334540	A	27-09-1989	AT 96126 T	15-11-1993
			AU 3167089 A	28-09-1989
			CA 1321711 A1	31-08-1993
			CA 2009641 A1	09-08-1990
			DE 68909979 D1	25-11-1993
			EP 0334540 A2	27-09-1989
			EP 0382442 A2	16-08-1990
			JP 1301501 A	05-12-1989
			JP 2790308 B2	27-08-1998
			NO 891272 A , B,	25-09-1989
			NZ 228400 A	26-07-1991
			US 5030661 A	09-07-1991
			ZA 8902181 A	31-01-1990
			AU 4929390 A	16-08-1990
			IN 176857 A1	21-09-1996
			IN 175181 A1	13-05-1995
			JP 2233502 A	17-09-1990
			NO 900608 A	10-08-1990
			NZ 232451 A	26-07-1991
			ZA 9000881 A	30-10-1991
US 4079017	A	14-03-1978	AR 226802 A1	31-08-1982
			BE 856919 A1	14-11-1977
			BR 7705012 A	06-06-1978
			CA 1084435 A1	26-08-1980
			DE 2741852 A1	24-05-1978
			ES 463927 A1	01-12-1978
			FR 2371377 A1	16-06-1978
			GB 1550754 A	22-08-1979
			IT 1083034 B	21-05-1985
			JP 1475287 C	18-01-1989
			JP 53064202 A	08-06-1978
			JP 62061521 B	22-12-1987
			MX 4298 E	18-03-1982

EPO FORM P0459

For more details about this annex : see Official Journal of the European Patent Office, No. 12/82

**ANNEX TO THE EUROPEAN SEARCH REPORT
ON EUROPEAN PATENT APPLICATION NO.**

EP 00 12 5077

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on
The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

07-02-2003

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
US 4079017	A		NL 7700934 A ,B, US 4162290 A	23-05-1978 24-07-1979
EP 0326662	A	09-08-1989	DE 3803082 A1 DE 3879924 D1 EP 0326662 A1 ES 2039561 T3	17-08-1989 06-05-1993 09-08-1989 01-10-1993
EP 0440258	A	07-08-1991	DK 28390 A AT 113260 T AU 624759 B2 AU 7017391 A CA 2035331 A1 CN 1056850 A ,B CS 9100248 A2 DE 69104735 D1 EP 0440258 A2 ES 2065556 T3 JP 1954871 C JP 4215837 A JP 6075670 B SK 279079 B6 RU 2053957 C1 US 5167933 A ZA 9101771 A	03-08-1991 15-11-1994 18-06-1992 08-08-1991 03-08-1991 11-12-1991 13-08-1991 01-12-1994 07-08-1991 16-02-1995 28-07-1995 06-08-1992 28-09-1994 03-06-1998 10-02-1996 01-12-1992 27-11-1991
US 3958951	A	25-05-1976	BE 827222 A1 CA 1078615 A1 DE 2513499 A1 ES 436042 A1 FR 2274672 A1 GB 1516361 A GB 1516362 A IT 1032412 B JP 1304508 C JP 50137888 A JP 60018601 B NL 7502563 A ZA 7501872 A	16-07-1975 03-06-1980 30-10-1975 01-01-1977 09-01-1976 05-07-1978 05-07-1978 30-05-1979 28-02-1986 01-11-1975 11-05-1985 13-10-1975 25-02-1976

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